Supporting Information

Experimental section

Materials
Kevlar 49 yarns were purchased from Dupont and used after dried at 50 °C in vacuum for 12 h. Dimethyl sulfoxide (DMSO), potassium hydroxide (KOH), polyvinyl alcohol (PVA), aniline monomer, and ammonium persulphate (APS), 12M (mol/L) HCl were purchased from Sinopharm Chemical Reagent Co. Ltd. All reagents were of analytical grade and used as received without further purification.

Preparation of ANFs-PVA Hydrogels
Aramid nanofibers (ANFs)/DMSO dispersion were prepared according to the method reported in the reference. Typically, bulk Kevlar 49 yarns (6.0 g) and KOH (9.0 g) were added to DMSO (300 mL) and stirred vigorously at room temperature for 3 weeks to obtain 20 mg/mL ANFs/DMSO dispersion. Then, 5 mL of ANFs/DMSO dispersion was mixed with an equal volume of a 100 mg/mL PVA (Mw 145000–165000 a.u.) solution in DMSO. The mixture system was cast into a mold and immersed in water to form ANFs/PVA hydrogels with the desired shape.

Preparation of APP Hydrogels
The ANFs-PVA hydrogel with a size of 1 × 3 × 0.03 cm$^3$ was immersed in 5 mL of 1 M HCl aqueous solution containing different initial concentrations of aniline monomer, and kept at 0 °C for 12 h. APS (mole ratio to aniline = 1:1, dissolved in 5 mL of 1 M HCl), pre-cooled to 0 °C, was poured into the above mixture, and the mixture was polymerized in situ at 0 °C. Finally, the composite hydrogel was thoroughly washed by HCl aqueous solution, ethanol and deionized water, respectively. According to the loading amount of polyaniline of 9.2 wt%, 14.7 wt%, 18.1 wt%, and 20.6 wt% (relative weight ratio of PANI to ANFs-PVA-PANI (APP) hydrogels), APP hydrogels (85% water content) were labeled as APP-0, APP-1, APP-2, APP-3 and APP-4, respectively.

Assembling of Strain Sensors Utilizing APP Hydrogels
To obtain the strain sensor, the excess water on the surface of APP-3 hydrogel (1 × 3 ×
0.05 cm³) was absorbed with filter paper, then two copper plates attached with conductive silver paste were connected to the both ends of the hydrogel (Figure 1).

![APP conductive hydrogel](image)

App. conductive hydrogel

Silver paste

Copper electrode

**Fig. S1** Three-dimensional structure of strain sensors assembled from APP hydrogels.

**Characterizations**

The fracture surface morphology of freeze-dried hydrogels was observed with field emission scanning electron microscopy (FE-SEM) (FEI-QUANTA250FEG, USA). Fourier transform infrared (FTIR) spectra were recorded on a Nicolet 8700 spectrometer (Thermo Fisher Scientific, USA) at a scanning resolution of 2 cm⁻¹ in the wavenumber range from 4000 to 500 cm⁻¹. Raman spectrometer (Via-H31894, Renishaw, British) were performed in the wavenumber range from 400 to 2000 cm⁻¹. X-ray diffraction (XRD) patterns were recorded on a D8-Advanced X-ray diffractometer (Bruker Co. Ltd, Switzerland) with Cu Kα radiation (λ = 0.154 nm) from 5 to 45°, at a scanning speed of 5° min⁻¹. The oscillatory frequency sweep measurements were performed at a strain amplitude of 5 % with shear frequency in the range of 0.1–100 rad/s at 25 °C to determine the storage and loss moduli of the hydrogels. The tensile process of hydrogels was characterized using a universal tensile testing machine (CMT 4254, Shenzhen SANS, China) with a tensile rate of 1 mm min⁻¹ at ambient temperature. The conductivity of the hydrogel was tested by a digital source meter (Keithley 2400, Tektronix, U.S.A.), and the piezoresistive performance was also tested by it combining with a universal tensile testing machine. The digital source meter was applied to evaluate performance of APP hydrogel sensors in practical applications.
For ANFs, the characteristic bands at 3313 cm\(^{-1}\) are attributed to the N-H stretching vibrations.\(^2\) The bands at 1631 and 1537 cm\(^{-1}\) are ascribed to the C=O stretching vibration and the N-H deformation vibration of amide group, respectively.\(^3\) The bands at 1501 and 1309 cm\(^{-1}\) are assigned to the C=C stretching and Ph-N stretching of benzene ring, respectively.\(^4\) The characteristic peaks of PVA are presented for the ANFs/PVA hydrogels. The peaks at 3286, 2904, and 1083 cm\(^{-1}\) are attributed to the vibrations of O-H, C-H, and C-O bonds of PVA, respectively.\(^5,6\) In ANFs-PVA composite materials, the C=O characteristic peak of ANFs obviously moves to the low band, which proves the existence of hydrogen bond between ANFs and PVA components.\(^2\)

![Fig. S2 FTIR spectra of ANFs, PVA and ANFs-PVA](image)

X-ray diffraction spectroscopy (XRD) scans of PVA, ANFs, PANI and APP hydrogels are shown in Figure S3 (Supporting Information). In the X-ray diffraction curve of PVA, the crystalline peaks appear at 2\(\theta = 19.5^\circ\), corresponding to the (101) plane of the PVA.\(^6\) The ANFs shows three typical Kevlar characteristic peaks, which
can be assigned to the (110), (200), and (004) diffractions, respectively.\(^2\) For the PANI, two broad peaks at around \(2\theta = 20.6\) and \(25.1^\circ\) are ascribed to the periodicity parallel and perpendicular to PANI chains.\(^3\) In the case of APP, the main characteristic peak was consistent with ANFs, but the intensity decreased significantly, which may be attributed to the introduction of the PVA and PANI.\(^4\)

![Fig. S3 XRD spectra of PVA, ANFs, PANI and APP.](image)

Fig. S3 XRD spectra of PVA, ANFs, PANI and APP.

![Fig. S4 (a) The stretching cycle at 60% strain. (b) The stretching cycle at 100% strain.](image)

Fig. S4 (a) The stretching cycle at 60% strain. (b) The stretching cycle at 100% strain.
References


